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Application of Environmental Isotopes as a Test
for Fracture Flow in Argillaceous Glacial Sediments

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APPLICATION OF ENVIRONMENTAL ISOTOPES AS A TEST FOR FRACTURE FLOW IN ARGILLACEOUS GLACIAL SEDIMENTS

By

Gregory Scott Foote

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ABSTRACT

APPLICATION OF ENVIRONMENTAL ISOTOPES AS A TEST FOR FRACTURE FLOW IN ARGILLACEOUS GLACIAL SEDIMENTS

By

Gregory Scott Foote

The effect of fractures upon the flow of solute through argillaceous glacial deposits in south-eastern Michigan was looked at by comparing measured concentrations of tritium from a continuous soil boring and from monitoring wells to simulated concentrations. The simulated values were determined by solute transport modeling of the tritium input function assuming matrix flow only (i.e. ignoring the impact of fractures). In addition, age, origin, and relationship to other Michigan groundwater was determined by comparing the del ^{2}H/ del ^{18}O ratio to that found by other researchers in Michigan.

The measured tritium distribution could not be simulated assuming matrix flow only. It was therefore concluded that fractures are an important mechanism of solute transport. The groundwater was determined to be modern in origin and isotopically similar to other groundwater sampled in Michigan.

ACKNOWLEDGEMENTS

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In addition, assistance was provided by colleagues John Gobins, Steve Young, and David Westjohn.

Jerry Fore, Wayne Disposal Inc., and RMT consulting are thanked for supplying the continuous soil boring B239 as well as for their cooperation during the course of this study. Bob Drimmie and the Environmental Isotope Laboratory at the University of Waterloo are thanked for their services.

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INTRODUCTION

The movement of ground water in non-indurated glacial sediments has been a topic of considerable research in recent years. This has been particularly true with respect to till and lacustrine clay deposits that cover vast regions of both the United States and Canada (see Lloyd, 1983 for review).

In the last few years, however, it has been proposed that the transport or movement of groundwater contaminants through argillaceous glacial sediments might be strongly influenced by the presence of fractures or other structure present in the sediment in addition to the matrix flow mechanism (Grisak and Jackson, 1974; Grisak, 1975; Grisak and Cherry, 1975; Williams and Farvolden, 1969; Hendry, 1982; Prudic, 1982; Sharp, 1984; Bradbury, 1984). This has led to much concern, particularly in the Great Lakes region, where argillaceous glacial deposits have been viewed as potential natural barriers to surface derived contaminants (Desaulniers et al., 1981).

Currently, only two studies focusing on the role of fractures in argillaceous glacial sediments have been made within the Great Lakes region (Desaulniers et al., 1981, 1982; Bradbury, 1984). However, to date, no studies have been undertaken within the State of Michigan to quantitatively determine what effect fractures might have on the migration of contaminants through these types of deposits. This is quite remarkable, since almost all shallow waste disposal facilities within the state are in argillaceous glacial deposits.

The focus of this research is to determine what effect, if any, fractures have upon the transport of solute (contaminants) through argillaceous glacial sediments located in southeastern Michigan. This study will use flow and solute transport modeling to simulate the distribution of tritium with depth assuming matrix flow to be the only component of flow. The simulated tritium distribution will then be compared to the actual to determine if fracture flow is an important component of flow. In addition, origin and relationship to other groundwater in Michigan will be determined by analyzing the groundwater present at the study area for the stable isotopes ^{18}O and ^{2}H.

STUDY AREA

The study area for this investigation is located in Augusta Township approximately 3.2 kilometers east of the town of Milan in Washtenaw County Michigan (Figure 1). The owners of this land, Augusta Development corporation (ADC), are projecting to use 1.62 of the 7.28 sq. km for a hazardous waste landfill facility. Prior to ownership by ADC, a parcel of land about .2 km west of the proposed

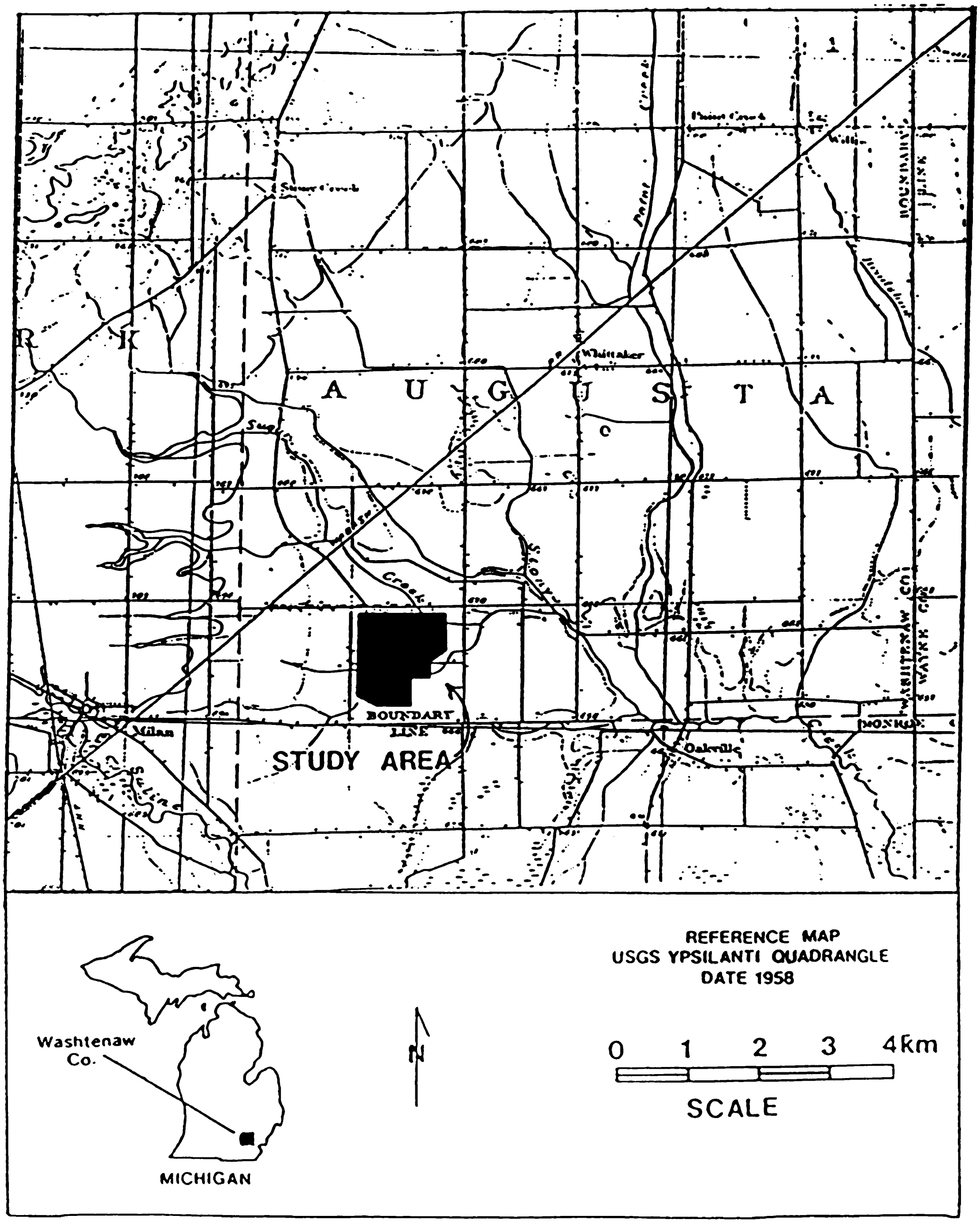


Figure 1. Geographic study area location map (contour interval = 20 ft.).

disposal site location had been used as a solid waste landfill (ADC Project Summary, in review). This facility stopped operation in the winter of 1979. Any cleanup of the old landfill required by the Michigan Department Of Natural Resources (MDNR) will be performed by ADC prior to its use as a hazardous waste facility.

The MDNR requires that a full hydrogeologic investigation to determine the suitability of the site be undertaken prior to its issuance of an operating permit. RMT Engineering and Environmental Management Services, Inc. of Madison, Wisconsin was employed by ADC to undertake this investigation. RMT has completed over 300 soil borings and 120 monitoring well installations, 26 of which are nested wells (Figure 2). Borings 163, 204, 239, 253, 332, 377, 501 and 502 are continuous down to bedrock . In addition, hydraulic conductivity, groundwater chemistry, soil grain size, and other analyses were determined by RMT.

GEOLOGY

The proposed Augusta hazardous waste facility (AHWF) lies on a lake plain associated with glacial Lake Warren (Farrand and Bell, 1982; Figure 3). The surface varies in elevation from 203 to 215 meters above sea level and slopes approximately .005 m/m towards the south east.

The sediments that underlie the facility range in thickness from approximately 28 to 40 meters and are shown

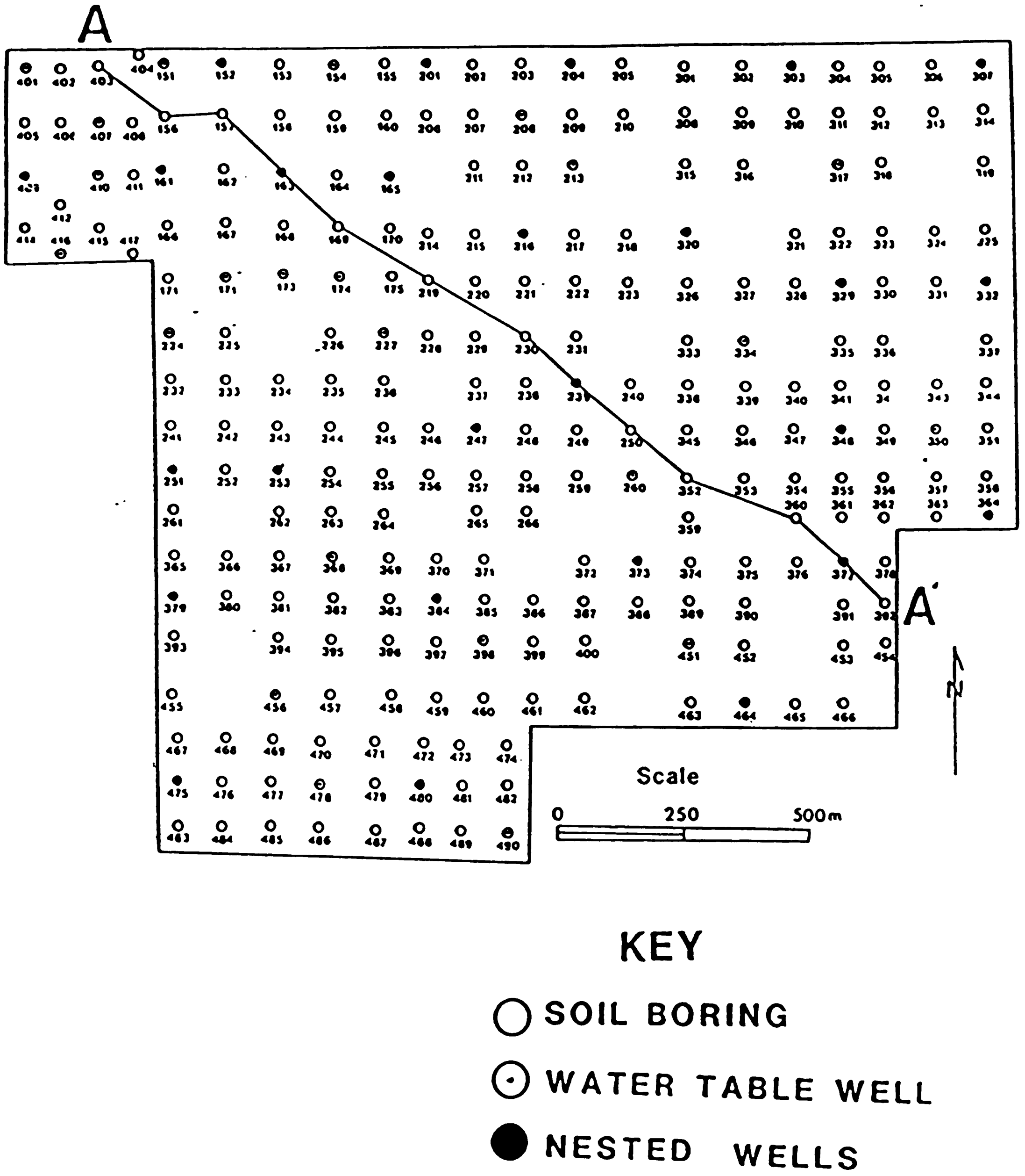


Figure 2. Soil Boring, nested well, and water table location map.

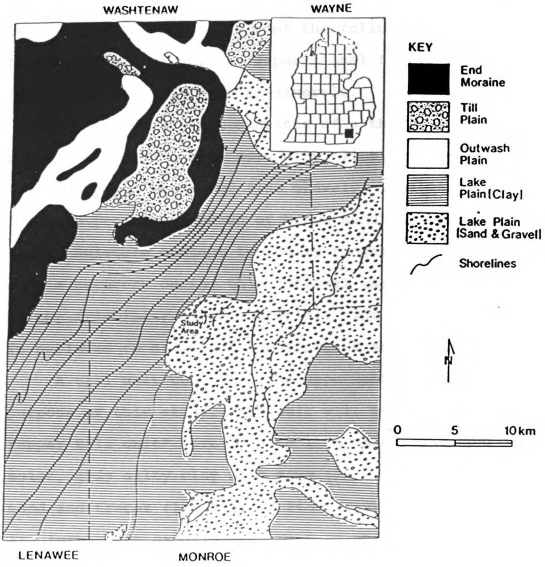


Figure 3. Geologic map showing location of study area and surficial deposits of Washtenaw, Wayne, Lenawee, and Monroe Counties (after Farrand and Bell, 1982).

in figure 4. These deposits can be broken up into four distinct units on the basis of the following parameters: grain size distributions, depositional features, and 7/10 angstrom ratios of the clay minerals.

The uppermost unit consists of a thin, .5 to 4.6 m thick, medium sand with some clay, little (12 - 23%) silt, and occasional traces of fine gravel. This unit grades downward from light brown or brown sand into gray silty sand. Moisture content typically ranges from moist to wet. Sample recovery during this interval was generally low due to the friability of the material. The origin of this unit is believed to be lacustrine (Farrand and Eschmann, 1982).

A till unit underlies the uppermost sand unit. For the purpose of this study, this unit will be referred to as the "upper till unit". It is 8 to 11 m thick, and consists of moist, gray silty clay with some (23 - 33%) fine-to-medium sand and traces (1 - 12%) of fine gravel. In addition there are infrequent sand lenses, most of which are found towards the north east. In an excavated pit approximately .25 km east of the AHWF, abundant fractures can be seen in the upper 3 to 5 meters of the upper till unit. These features are typically vertical, with aperture spacing ranging up to 5 cm in width. The fracture spacing is approximately 15 to 20 cm. Many of the fractures are coated with a brownish-yellow oxidation stain similar to that found in fractured tills by other investigators (Grisak, 1975; Grisak and

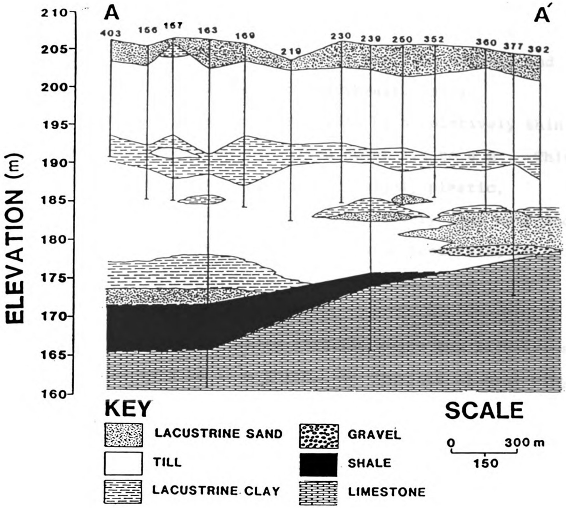


Figure 4. Geologic cross section showing subsurface geology along line A - A' of Figure 2. All elevations in m.a.s.l.

Cherry, 1975; Fookes, 1965; Eyles and Sladen, 1981; Kazi and Knill, 1973). The origin of this unit is believed to be from basal meltout of the Huron/Erie ice lobe (Morner and Dreimanis, 1973; Dreimanis and Goldthwait, 1973).

The upper till unit is underlain by a relatively thin clay that ranges in thickness from about 1.5 to 8 m . This unit consists of moist, gray, silty, highly plastic, laminated clay with occasional traces of very fine sand and fine gravel. This unit is believed to be lacustrine in origin (Morner and Dreimanis, 1973; Dreimanis and Goldthwait, 1973).

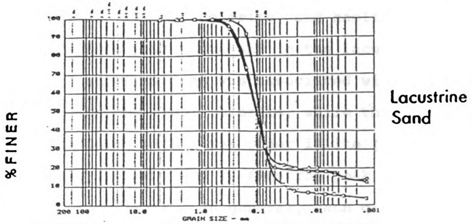
A "lower till unit" lies directly below the lacustrine clay unit and is 8 to 17 m thick. It consists of gray clay and silt with some fine to medium sand and trace fine gravel. In addition, there are abundant sand, gravel, and laminated clay lenses located in this layer. Most of the sand lenses encountered are relatively small and discontinuous; however, there are a few that are 300 to 400 m in length. These large sand bodies are generally found at the base of the till bedrock contact. Occasional "boulder zones" are also found at the lower till bedrock contact. This unit is believed to be from subaquaeous or flotation origin due to the large "pockets" of lacustrine clay intermixed with the till (Morner and Dreimanis, 1973; Dreimanis and Goldthwait, 1973).

Grain size distributions for each of the major glacial and lacustrine units are shown in figure 5. This data was obtained from sieve analyses of sediment derived from boring 239 and shows that the lowest three glacial units have distinctly different grain size distributions. The data also shows that both till units have a greater sand content than the lacustrine clay unit.

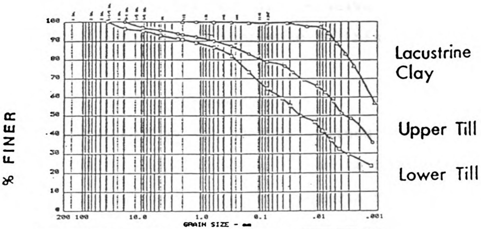
The Traverse Group underlies the glacial deposits at the AHWF. It is Devonian in age and consists of shale and limestone lithologies (Fleck, 1980). The uppermost unit is a gray, moderately hard, weathered, moderately fractured shale 0 to 14 m thick, which contains many fossils and some pyrite nodules. This unit pinches out towards the east. Underlying the shale is a gray, hard, massive, variably fractured limestone unit which contains fossils, vugs, and calcite crystals. An oily smell was reported in the borings that penetrated into the limestone unit and is probably due to the presence of methane.

CLAY MINERALOGY

Twenty four samples taken at various depths at the AHWF were analyzed by x-ray diffraction for clay mineral identification by Dr. Michael Velbel of Michigan State University's Geology Department. All samples underwent the following treatments: potassium saturation, magnesium/glycol saturation, and thermal heating to 550



% +3"	% GRAVEL	% SAND	% SILT	% CLAY
0.0	0.0	68.7	13.3	18.0
0.0	0.0	67.6	15.4	17.0
0.0	0.0	68.2	26.4	5.4



% +3"	% GRAVEL	% SAND	% SILT	% CLAY
0.0	4.5	16.5	22.6	56.4
0.0	0.0	0.6	9.2	90.1
0.0	7.1	28.1	30.4	34.4

Figure 5. Grain size distribution of sediment derived from soil boring number 239.

degrees celsius. Illite, chlorite, and kaolinite were identified in both till units and in the lacustrine clay layer. The 7 angstrom peak was associated with first order kaolinite and second order chlorite, while the 10 angstrom peak was associated with first order illite (Brown and Brindley, 1980). The 7 and 10 angstrom peak heights were compared for each layer. In general, each glacial and lacustrine unit showed a distinct 7 to 10 angstrom ratio signature (Table 1). However, the standard deviation of the lacustrine clay was consistently larger than that of either till unit. This indicates that the relative abundances of kaolinite and illite varied much more in this layer.

HYDROGEOLOGY

Surface runoff at the AHWF is drained via Sugar Creek and Augusta Drain. Approximately 0.2 km to the east, there is an open pit which hydraulically connects the limestone unit with the surface. This pit was previously used by the Martin Marietta corporation as a limestone quarry.

In figure 3, the cross section A to A' lies parallel to the direction of groundwater flow. The lateral gradient ranges from nearly zero towards the center of the A to A' traverse to .0021 m/m towards the south east. A downward vertical gradient of approximately .74 m/m was calculated from the static-water elevations of the nested wells that are screened within the glacial, lacustrine, and bedrock

Table 1. 7/10 angstrom ratio data.

Treatment: Potassium saturation

UNIT	MEAN 7/10 A	Std. Dev.
Upper Till	.5845	.0371
Lacustrine Clay	.7689	.1293
Lower Till	.7115	.0277

Treatment: Magnesium/Glycol saturation

UNIT	MEAN 7/10 A	Std. Dev.
Upper Till	.7160	.0611
Lacustrine Clay	.8842	.1369
Lower Till	.8475	.0708

units (figure 6). A comparison of the lateral and downward hydraulic gradients indicates that the predominant direction of groundwater movement is downward.

The upper sand unit, the extensive sand lenses in the lower till unit, and the Traverse group limestone unit are the primary water bearing units at the facility.

Field and laboratory hydraulic conductivity tests were performed on each of the glacial and lacustrine units and on the limestone unit. The field hydraulic conductivity tests were performed using the Bouwer and Rice and/or the Hvorslev method of calculation (Hvorslev, 1951; Bouwer and Rice, 1976). Laboratory hydraulic conductivity tests were performed using the falling head permeameter method (Olson and Daniel, 1981). The results of these tests are summarized in table 2.

From the above data, it is apparent that the limestone unit displayed the largest variation in hydraulic conductivity (k). This can be explained by the wide range of fracture and vug densities that are encountered in the borings of this unit. In areas of extensive fracturing, or in the presence of a large number of vugs, the hydraulic conductivity would be expected to be high. No testing of the Traverse Group Shale was conducted; however, the hydraulic conductivity of this unit is assumed to be low. In general, the lower till unit demonstrates a higher k than the overlying lacustrine clay unit and upper till unit.

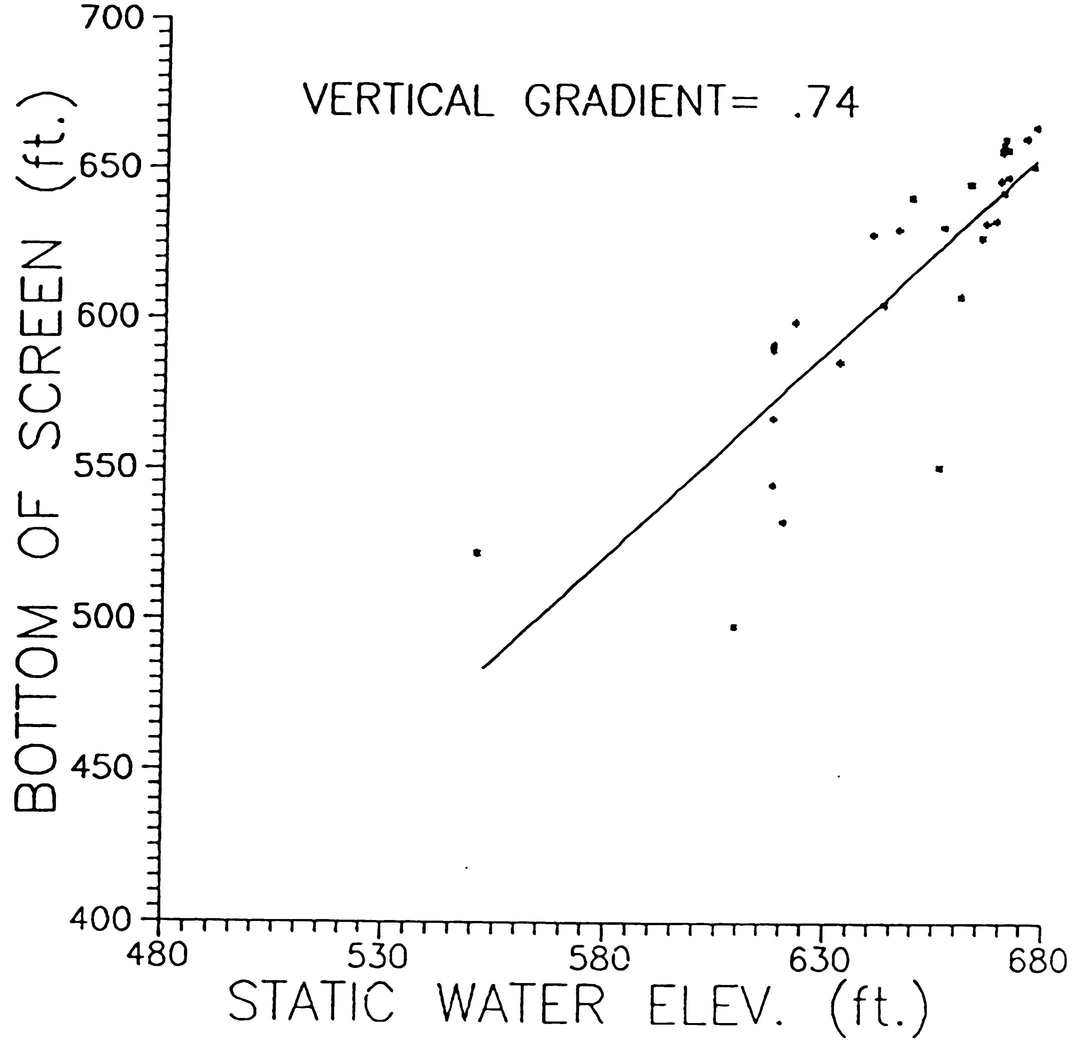


Figure 6. Elevation of static water level versus elevation of bottom of screened interval of nested and water table wells.

Table 2. Hydraulic Conductivity Summary

Geologic Unit	Field k Range (cm/s)	Laboratory k Range (cm/s)
Lacustrine Sand	5.5 * 10-4 to 1.69 * 10-6	8.4 * 10-8
Upper Till	1.4 * 10-6 to 3.0 * 10-8	3.0 * 10-8 to 1.3 * 10-8
Lacustrine Clay	4.9 * 10-8 (only 1 k test was run on this interval.)	N/A
Lower Till	3.5 * 10-5 to 2.4 * 10-7	4.2 * 10-6
Limestone	6.8 * 10-4 to 2.0 * 10-8	N/A

This reflects the numerous coarse textural heterogeneities found in the lower till unit. The lacustrine clay unit is the hydraulically tightest unit of the glacial sediments. Much of the hydraulic head loss observed in the nested wells was associated with this layer. In the upper till and sand units, it should be noted that the laboratory k values were all less than or equal to those reported in the field k testing. This is a common phenomena when large scale heterogeneities and/or structure (i.e. fractures) are present within the sediment.

NUMERICAL SIMULATION OF GROUNDWATER FLOW

The groundwater flow along cross-section A - A' was simulated using the three dimensional, finite difference flow model - MODFLOW (McDonald and Harbaugh, 1988). Two dimensional flow was assumed along cross-section A - A' since this traverse is located parallel to the regional groundwater flow direction and the primary direction of flow is downward.

In figure 7, the grid spacing for the flow model is shown. An 8 layer, 20 column, 1 row grid was used. The layer spacing was determined by grouping sediments of similar geologic characteristics into layers. The following layer spacing was used based upon the above constraint: layer 1= 2.88 m, layer 2= 5.76 m, layer 3= 4.61 m, layer 4=

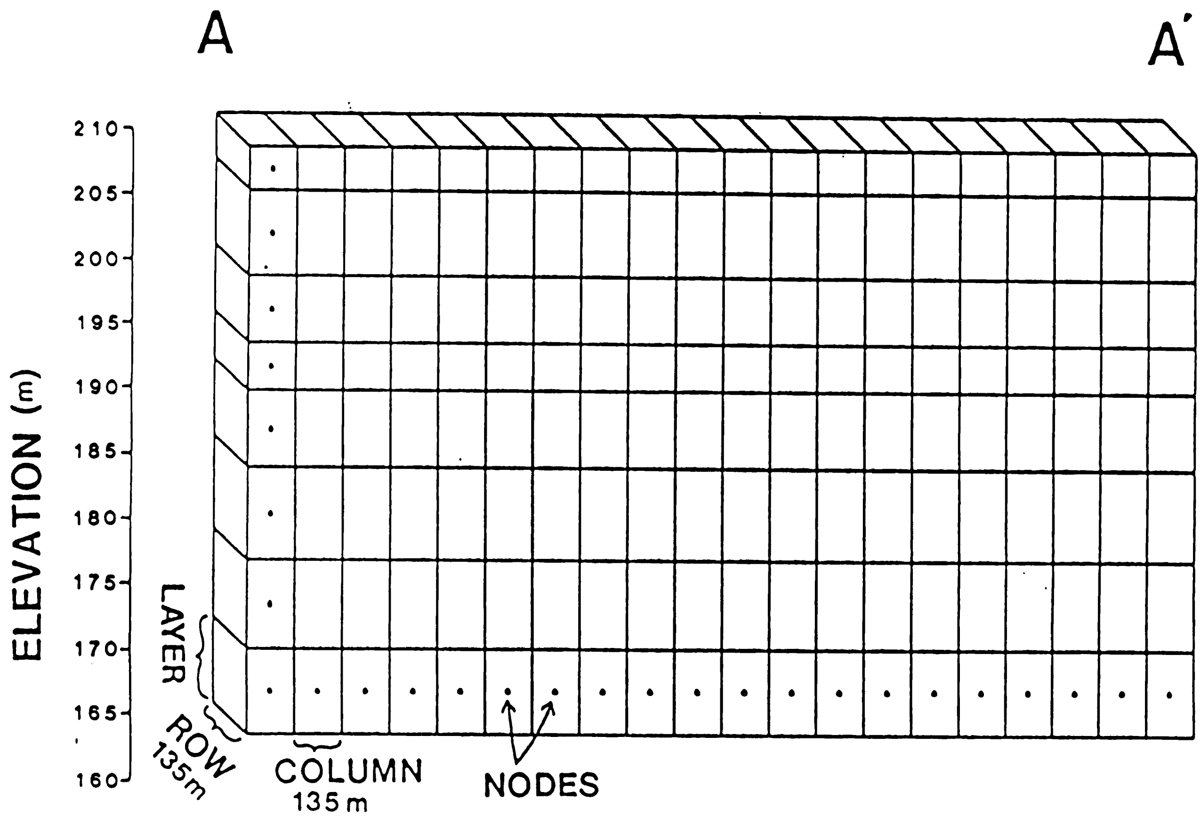


Figure 7. Flow model grid spacing. 1 row * 20 columns * 8 layers. Layer 1 represents upper sand unit, layers 2-3 represent the upper till unit, layer 4 represents the lacustrine clay unit, layers 5-6 represent the lower till unit, and layer 8 represents the bedrock.

3.26 m, layer 5= 5.18 m, layer 6= 6.34 m, layer 7= 5.95 m and layer 8= 5.95 m. There are a total of 160 nodes incorporated in the model (i.e. 8 layers x 20 columns x 1 row = 160 nodes).

Prior to running a model simulation, the initial parameters of confining, discharge and starting head conditions were determined for each node. The starting head values were estimated based on the static water levels observed in observation wells. In locations where there were no wells, two point linear interpolation between adjacent water levels was used. The confining conditions were determined based on the hydraulic conductivity of the layer and its location relative to low k confining layers. From this definition the uppermost layer was modeled as unconfined, while the underlying seven layers were modeled as confined. Since the primary direction of groundwater movement along cross section A - A' is downward through the glacial and lacustrine sediments into the bedrock, it was necessary to simulate this groundwater movement using a series of discharge wells. Discharge wells were located at each node in layer 8, the bedrock layer. The rate of discharge was determined by the following relationship:

Eq. 1 Discharge (m^3/day)= Column width (m) x Row width (m) Vertical hydraulic conductivity (m/day) x Vertical anisotropy x Vertical gradient

The discharge parameter varied across the bedrock layer due to the change in hydraulic conductivity between the shale and limestone that occurs between columns 6 and 14 or the variation in the hydraulic conductivity of the limestone. The discharge rate was larger in the limestone than in the shale due to the greater hydraulic conductivity of the limestone. In the shale-to-limestone transition zone the hydraulic conductivity was determined by spatially calculating an aeriel percentage of each sediment type present, multiplying this by its respective k and then summing the products to come up with a weighted average k.

Once all the initial parameters were determined, the model was calibrated by adjusting the conductivity of several of the nodes until the simulated heads were consistent with those observed. All of the k values were kept within the range of values reported by the slug and bail tests (Table 2). Slight adjustments were also made to the pumping rates of the discharge wells.

The model was initially set to calculate its own recharge rate. This was accomplished by defining the upper layer as constant head, which allowed the model to determine how much recharge was needed to keep the upper layer heads at their starting elevation. Once the model was calibrated, the upper layer constant head condition was replaced by one of variable head. The amount of recharge that the model calculated as necessary was then added onto the upper layer of the model. This value, 5.59 cm/yr (2.3 in/yr) was within the range of recharge values (5.08 - 10.9 cm/yr) calculated by the baseflow separation of nearby Stoney Creek (United States Geological Survey, 1980).

The resultant simulated heads are shown as equipotential lines in figure 7 . From the simulated heads, it is evident that the movement of the groundwater throughout the AHLF is primarily downward except in the lower till above the shale to limestone transition zone, where the direction of flow is towards the south east (i.e. towards the limestone that is "exposed" by the shale pinchout). It should also be noted that the greatest amount of head loss is associated with the lacustrine clay unit. In addition, the vertical gradient through the upper till and lacustrine units is .76 m/m. This value agrees closely with the average vertical gradient calculated from static water elevations from the observation wells (Figure 6).

The average flow velocity in the lacustrine sand and upper till can be calculated from the following Darcian relationship:

$$\text{Eq. 2} \quad V = ki/n$$

where- k= hydraulic conductivity (m/day), i= vertical gradient, and n= porosity. Therefore, for the lacustrine sand, V= .0864 m/day * .28 / .35 = .0691 m/day, and for the

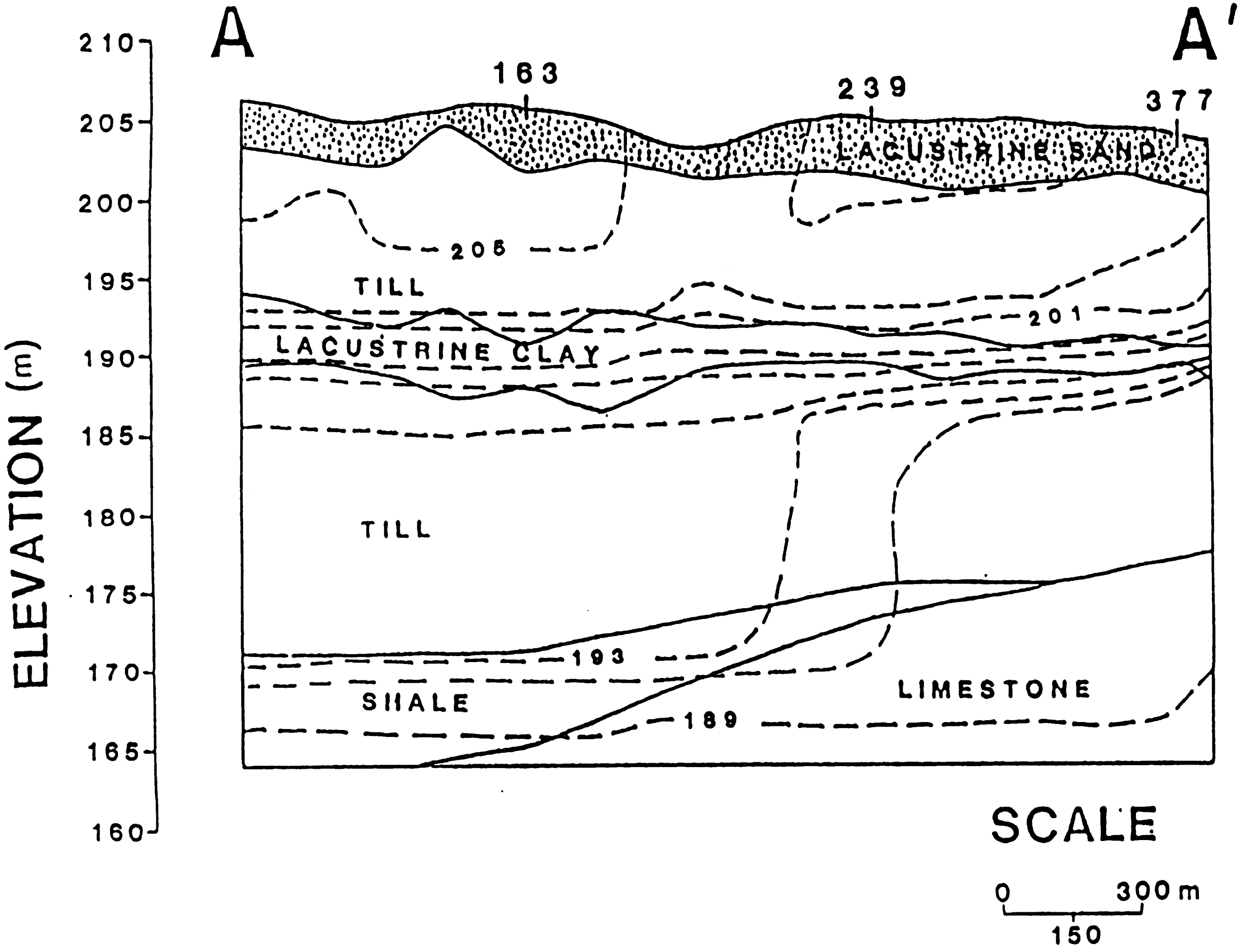


Figure 8. Simulated heads contoured as equipotential lines (contour interval = 2 m).

upper till, V= 4.62*10-5 m/day * .76 / .30 = 1.17*10-4 m/day. This is based on a calculated average hydraulic conductivity from the field for each unit, estimated average porosity values based on sediment type (Fretter, 1988), and on the average hydraulic gradient for each layer as derived from the flow model.

ENVIRONMENTAL ISOTOPES

Soil boring 239 was supplied by RMT with permission from ADC and was chosen for soil water extraction because it is continuous to bedrock, is centrally located on the site, has little in the way of missing sample intervals, and is also a good representative sample of the site-wide geology. From this boring, 17 water samples were extracted at .5 to 2 m intervals by azeotropic distillation (Figure 9; Appendixes A and B). The samples derived from this method were tightly sealed and delivered to the University of Waterloo Environmental Isotope Laboratory for isotopic analysis. Each of these samples were analyzed for tritium by direct liquid scintillation counting. This method yielded a detection limit of 6 TU (tritium units, where 1 TU= 1 tritium atom per 10^{18} hydrogen atoms) and a counting error of +/- 8 TU.

In addition to the extracted samples, groundwater samples were collected from well nests 163, 239, and 377. For each well nest there is a total of 5 wells screened at

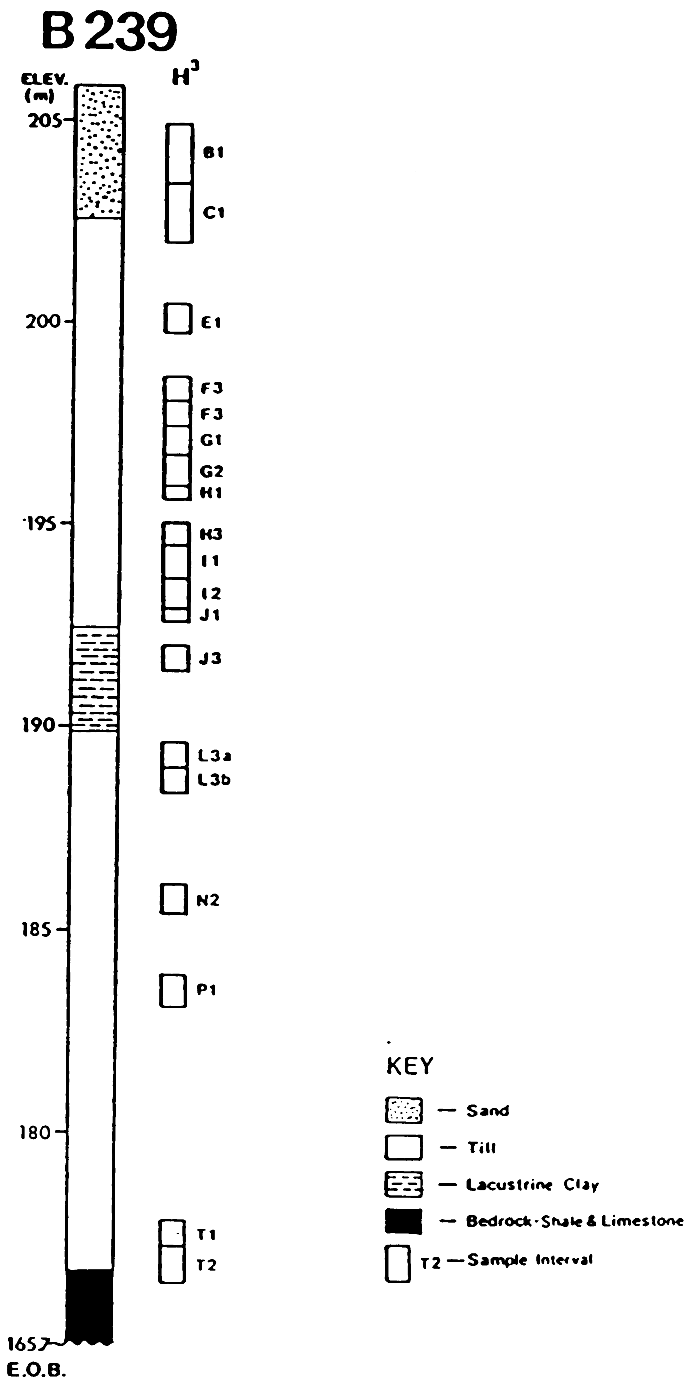


Figure 9. Soil boring 239, location of samples obtained by azeotropic distillation.

various intervals (Table 3). To insure an uncontaminated representative sample, each well was purged of 3 to 5 well volumes the day before sampling. A sample was taken from each of the wells with the exception of 377A, which was dry at the time of sampling and 163B, 239B, and 377B, which were later sampled by RMT. All samples were collected with a teflon bailer and then decanted into 1l Nalgene sample bottles. In order to prevent exchange with atmospheric oxygen and hydrogen, all sample bottles were filled to the top and tightly sealed. Once the samples had been collected, they were electrolytically enriched at the Michigan State University Tritium Laboratory to decrease error involved with the liquid scintillation counting method (Wyerman, 1978; Gobins, 1989; Ostlund and Werner, 1962; Packard, 1987). The samples were then sent to the University of Waterloo Environmental Isotope Laboratory for isotopic analysis. The counting error for the enriched samples ranged from +/- .69 - 1.92 TU (Table 6).

Samples for stable isotope (^{2}H and ^{18}O) analysis were obtained from wells 239, 239A, 239C, and 239D. Each of these samples were collected in the same manner and at the same time as those obtained for the tritium analyses. All samples were sent to the University of Waterloo Environmental Laboratory for stable isotope analysis by mass-spectrometry. The accuracy of this method is +/- 0.2 0/00 for ^{18}O and +/- 2 0/00 for ^{2}H.

Table 3. Nested well sample interval elevation and geologic unit sampled.

WELL ID	SAMPLE INTERVAL (m)	GEOLOGIC UNIT
163	205.4 - 202.4	LACUSTRINE SAND
163A	199.9 - 198.4	UPPER TILL
163B	194.3 - 192.8	LACUSTRINE CLAY
163C	185.9 - 184.3	LOWER TILL
163D	165.4 - 162.3	LIMESTONE
239	204.2 - 201.2	LAC. SAND/ UPPER TILL
239A	198.8 - 197.2	UPPER TILL
239B	194.1 - 192.5	U. TILL/LAC. CLAY
239C	185.6 - 182.5	LOWER TILL/ SAND
239D	169.0 - 166.0	LIMESTONE
377	203.5 - 200.5	LAC. SAND/U. TILL
377A	196.7 - 195.2	UPPER TILL
377B	192.9 - 191.4	LACUSTRINE CLAY
377C	181.8 - 180.3	SAND/ UPPER TILL
377D	175.7 - 172.7	LIMESTONE

STABLE ISOTOPE RESULTS

Table 4 is a summary of the stable isotope results. Figure 10 shows the del ^{18}O versus del ^{2}H isotopic values from this study and from other groundwater in Michigan as well as the Lake Simcoe, Ontario meteoric line. From the data in figure 10, it is evident that analyses from the AHWF are consistent with those found by other workers in Michigan (Long et al., 1988; Regalbuto, 1987; and Gobins, 1989). In addition, the data reflect a linear trend that is parallel to the Lake Simcoe trend but falls beneath the Lake Simcoe meteoric line. By running a linear regression on the Michigan data, a best fit line (i.e. Michigan meteoric line) was found to be:

Eq.3 $$\text{del }{}^{2}H = 7.22 * \text{del }{}^{18}O + .429$$

The data in figure 11 indicates that all the groundwater sampled at the AHWF are modern in origin, since they are undepleted relative to SMOW even in the deeper lower till and limestone units.

TRITIUM RESULTS

The results of the tritium analyses from soil boring 239 are shown in table 5 and the results of the tritium analyses from the nested wells are shown in table 6. Any

Table 4. Stable isotope results.

WELL ID	ELEVATION SAMPLE INTERVAL (m)	del ^{18}O	del ^{2}H
239	204.22 - 201.17	-8.85	-59.78
239A	198.76 - 197.24	-8.62	-58.25
239C	185.56 - 182.51	-8.52	-59.14
239D	168.86 - 165.96	-8.97	-64.16
	(sample duplicate)	-8.74	-59.84

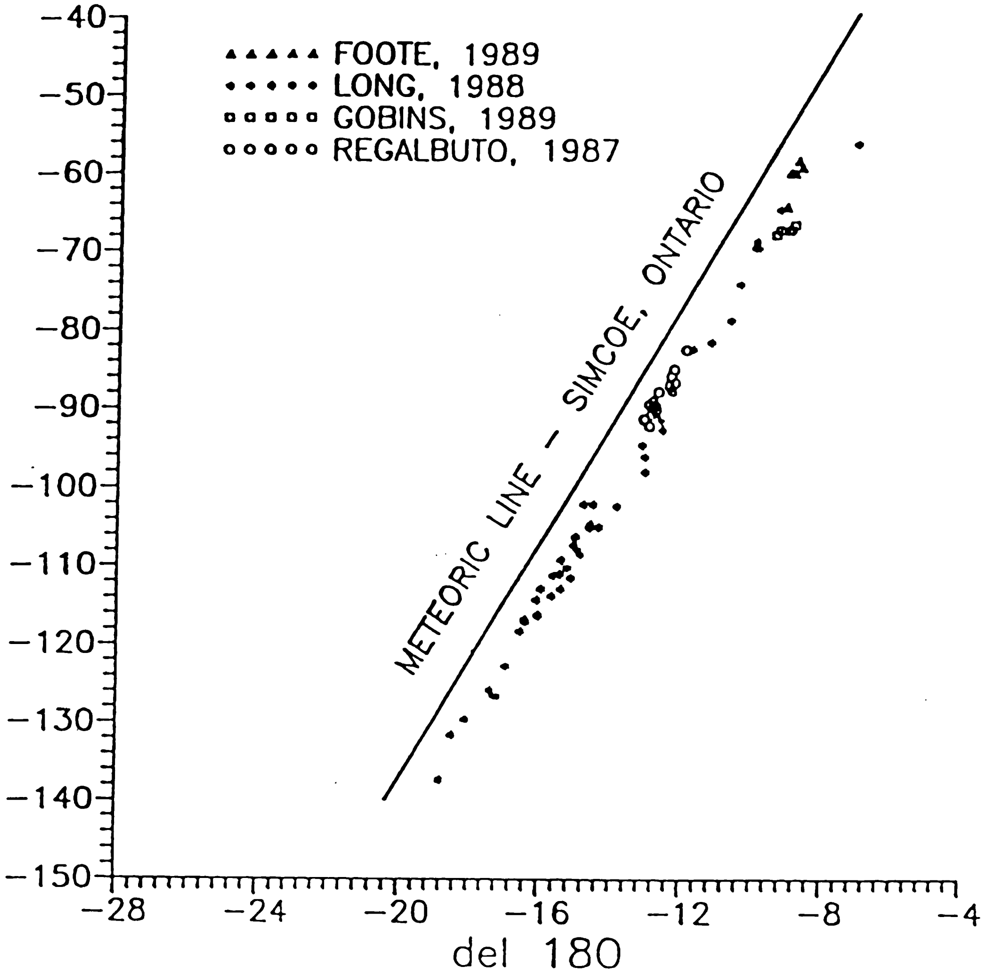
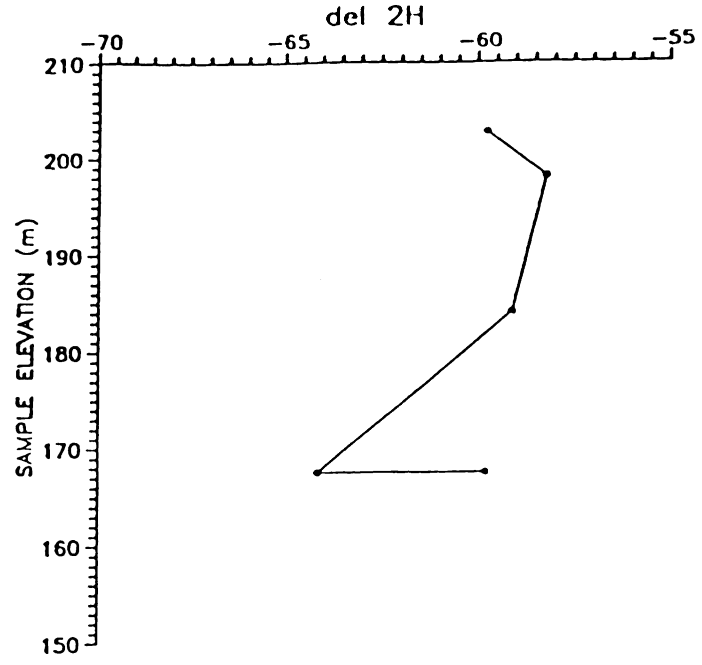


Figure 10. Stable isotope composition of water sampled from drift wells in Michigan.



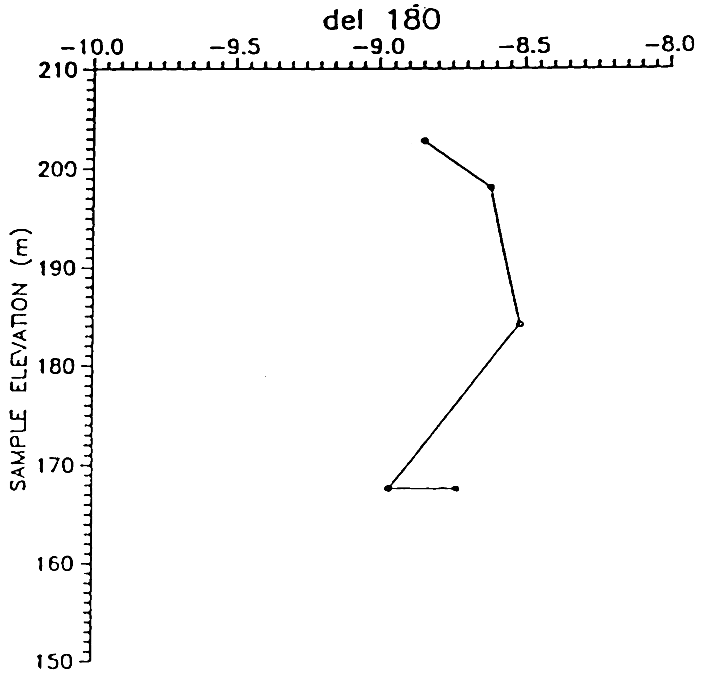


Figure 11. ^{2}H versus depth and ^{18}O versus depth for water sampled from wells 239, 239A, 239C, and 239D.

Table 5. Tritium results for extracted samples.
(note: detection limit = 6 TU and error = +/- 8 TU.)

SAMPLE ID	TU'S	SAMPLE INTERVAL (m)
B-1	28	.91 - 2.44
C-1	33	2.44 - 3.96
E-1	12	5.49 - 6.25
F-3	<6	7.92 - 8.53
G-1	6	8.53 - 9.30
G-2	12	9.30 - 10.06
H-1	<6	10.06 - 10.36
H-3	6	10.97 - 11.58
H-3 DUPLICATE	<6	
I-1	<6	11.58 - 12.34
I-1 DUPLICATE	8	
I-2	16	12.34 - 13.11
J-1	<6	13.11 - 13.41
J-3	<6	14.02 - 14.63
L-3	11	16.46 - 17.07
N-2	<6	19.96 - 20.73
P-1	8	22.25 - 23.01
T-1	<6	28.35 - 28.96
T-2	<6	28.96 - 29.87

Table 6. Tritium results for nested well samples.

WELL ID	TU'S	COUNTING ERROR
163	27.69	+/- 1.19
163A	2.39	+/- 1.06
163B	.21	+/- 1.92
163C	.67	+/- 0.69
163D	4.05	+/- 1.20
239	17.27	+/- 1.08
239A	7.66	+/- 1.07
239B	11.28	+/- 1.46
239C	3.09	+/- 0.90
239D	.74	+/- 1.05
377	11.40	+/- 1.04
377B	11.35	+/- 1.44
377C	2.31	+/- 0.87
377D	.63	+/- 1.12

sample that contains more than .45 TU of tritium above its counting error can be considered to contain "bomb tritium", and was therefore recharged prior to 1953 (Robertson and Cherry, In Press).

Figure 13 is a plot of tritium concentration versus depth for the extracted samples derived from soil boring 239. "Bomb" tritium was found at depths exceeding 22 m in these samples, while non-detectable values (<6 TU +/- 8 TU) were found as shallow as 8 m depth. Figure 14 shows the tritium concentration versus depth for the nested well samples. "Bomb" tritium is found at depths of at least 25 m in the glacial sediments, while groundwater with non-detectable tritium concentrations (1.5 TU +/- .69 - 1.92 TU) was found as shallow as 14 m. The nature of the tritium profile can be identified much more easily for the well samples as opposed to the extracted samples, since the counting error is much less due to the electrolytic enrichment process.

In both the extracted and nested well tritium profiles, there is a similarity in that, tritium "spikes" (i.e. relatively high tritium concentrations) are located intermediate in depth to non-detectable tritium concentrations.

It must be noted that, since fluids were used during the well drilling process, some artificial introduction of tritium into the sediment and bedrock may have occurred.

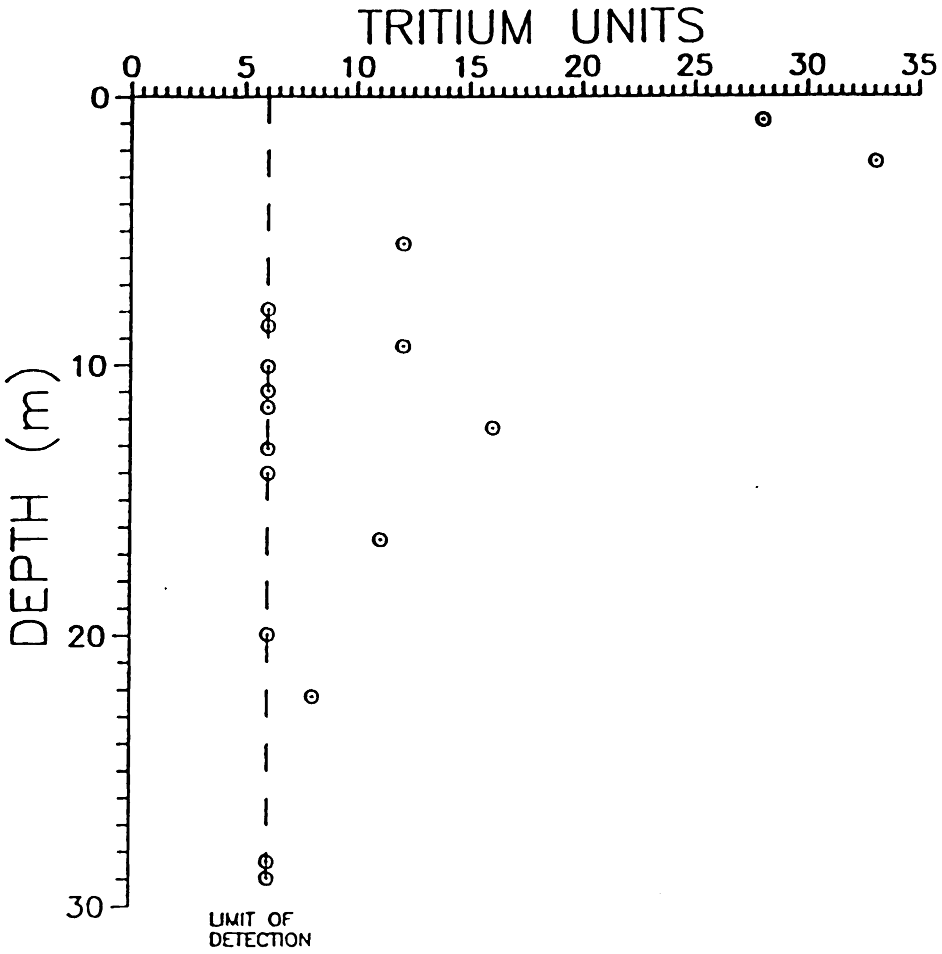


Figure 12. Tritium versus depth for extracted samples.

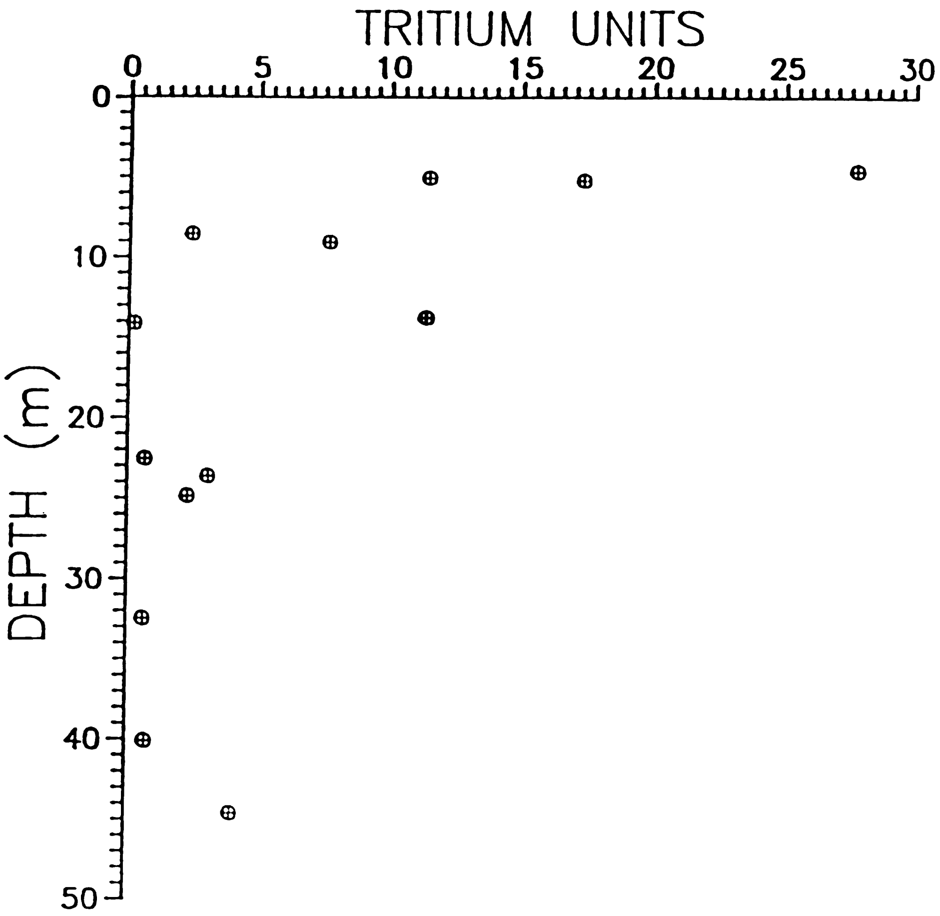


Figure 13. Tritium versus depth for nested well samples.

However, since the wells were purged prior to sampling and there is a close agreement between the extracted and nested well tritium values, this effect is considered to be minor.

DISCUSSION

The effect of hydrodynamic dispersion (D) on the flow of tritium through the lacustrine sand and upper till units at the AHWF was simulated using a one dimensional solute transport model (Javendal et al., 1984). This model takes into account advection, dispersion, and molecular diffusion based upon the following solution to the one dimensional solute transport equation:

Eq.4
$$D \frac{d^2C}{dx^2} - v \frac{dC}{dx} - LC = \frac{dC}{dt}$$

$$D = av + D*$$

where x is the distance travelled, t is the travel time, v is the average linear velocity in the x direction, C is the solute (tritium) concentration, L is the decay constant of the solute, and D is the dispersion, which is based upon the dispersivity - a, the linear velocity - v, and the molecular diffusion coefficient - D*. Assumptions inherent in the model are that groundwater movement is: exclusively by matrix flow, in the x direction (downward), and that the transport parameters (i.e. a, v, and D*) are constant in the direction of flow. The superposition method (Egboka et al., 1983) was implemented to account for variations in the tritium input over time. The tritium input function applied to the model was based upon yearly averages of tritium concentration from precipitation that fell from 1953 to present at Chicago, Illinois (Appendix E).

One problem that arose when applying the model at the AHWF was that the assumption that the flow parameters remained constant in the direction of flow was not met precisely as the tritium (solute) moves through the lacustrine sand unit down into the upper till unit. However, this limitation in modeling was overcome by first modeling the transport of the initial tritium input function through the lacustrine sand unit, and then modelling the resultant output of tritium through the base of the upper sand unit into the upper till unit. To accomplish this, the following transport parameters were used: a dispersivity value of .02 m for sand (Robertson and Cherry, in press), a linear flow velocity of .0691 m/day, and an assumed negligible D* value. The D* can be assumed to be negligible when the av term from equation 4 is large with respect to D* (i.e. when the linear flow velocity and/or the dispersivity are high)(Sternberg, 1985). Due to the rapid flow velocity and the relative thinness of this layer, there was no appreciable difference between the initial input function and the resultant modified tritium input function.

Assuming movement strictly by matrix flow, the following transport parameters were used to model the input of tritium into the upper till unit: a range of dispersivity values for argillaceous glacial sediments from .1 - 10 m (Ilgenfritz et al., 1988), a linear flow velocity of 1.17 * 10-4 m/day, and a D* of 4.32 * 10^{-6} m^2/day was used and is based upon research by Gilliam et al., (1984) on the effects of diffusion of non-reactive solutes in clay-rich sediments. The effects of molecular diffusion could not be considered negligible for the upper till unit due to the relatively slow linear groundwater velocity. The results of the solute transport simulations through the upper till are shown in figure 14.

If the movement of tritium through the upper till was solely by transport via matrix flow, then it should be possible to duplicate the distribution of the tritium in the till using one assumed dispersivity value. It is apparent, however, that none of the dispersivity values produce a simulation that adequately defines the tritium distribution within this unit. For example, the simulation in which the dispersivity was set equal to 1 m came close to duplicating the tritium peak at about 4 m depth, however, it cannot explain the presence of "bomb" tritium at depths beyond about 8 m. In addition, simulations which used a

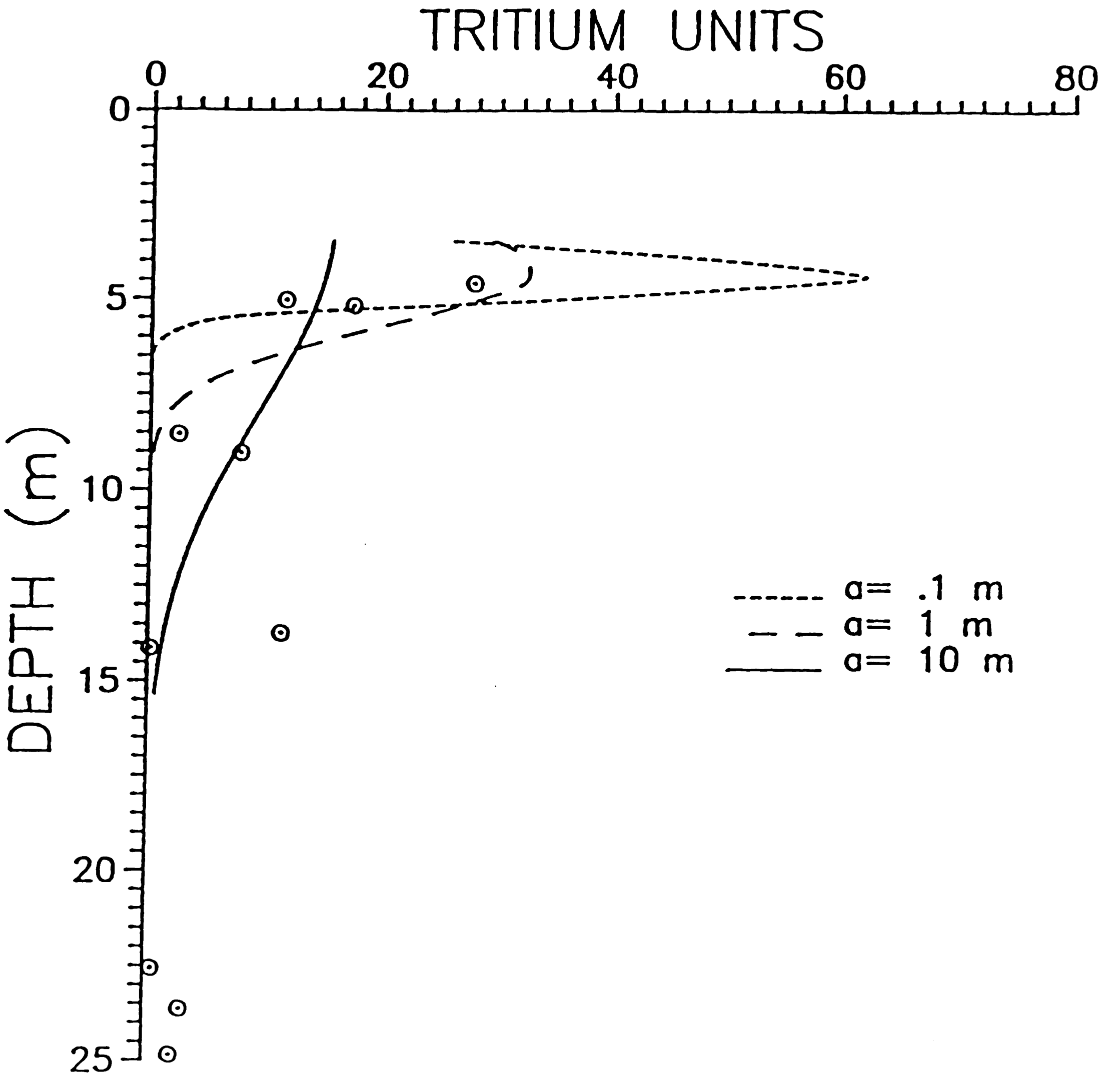


Figure 14. Simulated tritium distribution along with observed tritium distribution (within nested wells).

dispersivity much above 1 m were not able to duplicate the tritium peak towards the top of the upper till.

The inability of the model to duplicate the distribution of tritium within the upper till using any single dispersivity value, suggests that the assumption of only matrix flow is not valid, and that there exists a faster flow mechanism (i.e. fracture flow) in addition to matrix flow. Furthermore, the presence of "bomb" tritium intermediate in depth to non-detectable values in both the nested wells and in soil boring 239 also suggests the existence of fracture flow.

SUMMARY AND CONCLUSIONS

Groundwater samples were obtained from 17 different elevations of a continuous soil boring and from 15 nested wells for tritium analysis. Detectable concentrations of tritium were found as deep as 25 m, while non-detectable tritium concentrations were found as shallow as 8 m in the argillaceous glacial sediments that underlie the proposed Augusta Hazardous Waste Facility (AHWF). It was concluded that fracture flow is an important component of flow at the AHWF. Evidence for this is as follows: first, the tritium concentration versus depth profiles could not be simulated through modeling assuming matrix flow only; and second, "bomb" tritium was found intermediate in depth to non-detectable tritium concentrations in the tritium concentration versus depth profiles.

Groundwater origin and relationship to other Michigan groundwater was determined by obtaining 4 groundwater samples from different elevations of a nested well group and analyzing for the stable isotopes ^{2}H and ^{18}O. All of the groundwater sampled at the facility was determined to be modern in origin, and isotopically similar to other groundwater from Michigan.

APPENDICES

APPENDIX A

THEORETICAL CONSIDERATIONS OF
SOIL WATER EXTRACTION

The extraction of water from soils has been accomplished by a number of different methods, namely-pressure plate filtration, high speed centrifuging, air drying, oven baking and azeotropic distillation (Hendry, 1983; O'Neil and Kharaka, 1975). When determining which extraction process was most appropriate, a number of different factors had to be considered.

One of the key parameters is the relationship between the water molecule and the clay mineral surface. Grim (1953) sites the existence of three types of water each with a different association to the clay mineral surface.

Pore water is the first type of water considered by Grim. This type of water is found in the pores of the sediment, on the surfaces and around the edges of the clay particles. Pore water is not bonded to the clay mineral and is therefore highly mobile, free to move throughout the sediment. The result of this unbonded highly mobile state is that the pore water can be driven off completely by heating at low temperatures (<100 degrees C).

The next type of water that needs to be considered is interlayer water. Interlayer water is located between the clay particle layers of the hydratable clay minerals (e.g. smectite, vermiculite, halloysite, etc.). This type of water is bonded to the clay mineral by the attraction of the positively charged end of the polar water molecule to the negatively charged clay mineral surface. Several researchers have shown that this type of water is physically different than the non-oriented pore water (Yariv and Cross, 1979; Grim, 1953). Temperatures in excess of 100 degrees celsius are considered necessary to fully drive off interlayer water (Grim, 1953).

The third type of water discussed by Grim (1953) is structural or OH lattice water. This type of water is physically incorporated into the clay mineral structure in the form of hydroxyl groups. The bonds formed by these hydroxyl groups are much stronger than those formed by the interlayer water. This results in a relatively high temperature being required to dehydroxylate the clay mineral. Temperatures exceeding 300 degrees celsius are considered necessary by Grim (1953).

The above leads to the next critical factor which is: to what degree do the three types of water interact or exchange with one another? This relationship is critical since the type of water we wish to extract will be based upon how rapidly this exchange takes place. For example, if the recharging groundwater were to become tied up by adsorption into the clay mineral structure and interlayer water and if the rate of exchange were low, then this would limit the water of interest to strictly that of pore water since each of the water types would be different isotopically. This would pose a problem when using thermal extraction methods since there is an overlap in the temperatures required for liberation of the pore and interlayer waters. However, Savin (1967) and Stewart (1972) demonstrated that the exchange rate between the interlayer and the bulk pore water to be extremely rapid- on the order of a few hours. Furthermore, several researchers have shown that the structural water of clay minerals is reflective of formation under isotopic equilibrium with its formational environment (Savin and Epstein, 1970a; Savin and Epstein, 1970b; Lawerence and Taylor, 1972; O'Neil and Kharaka, 1975; Yeh and Epstein, 1978). This indicates that the rate of exchange between pore and interlayer waters with structural water occurs very slowly under sedimentary conditions.

The above discussion in conjunction with yield required and contamination concerns places the following constraints upon the extraction method:

1.) That a method must be chosen that can remove both the pore and interlayer waters while leaving the structural water intact. Therefore, if thermal methods are used, then the temperature must be carefully monitored to remain below the temperature of dehydroxilation.

2.) The volume of water extracted must be greater than 10 ml since at least this volume is required for the tritium analyses.

3.) No other fluids that contain water may be used in the process since this could cause sample contamination.

Several of the previously considered extraction methods were eliminated on the basis of the above considerations. For example, the high speed centrifuge method was eliminated due to low yield, the pressure plate filtration method for risk of sample contamination and several of the thermal methods were thrown out due to sample collection problems and the relatively high temperatures that were needed in order to attain a high yield.

The azeotropic distillation method was chosen because it fit the above criteria and it had been used successfully in a similar study (Hendry, 1983). For a discussion of the methodology of this technique see appendix B.

APPENDIX B

AZEOTROPIC DISTILLATION
LABORATORY METHOD

The laboratory setup for azeotropic distillation is a slight modification of that used by Hendry (1983) (figure 15). The only piece of equipment that had to be especially made was the buret stopcock connector which served to connect the condenser to the boiling flask and to collect the extracted water. This device was constructed by the Michigan State University glass blowers.

The laboratory method is as follows:

1.) Approximately 300-1000 gm of soil was disaggregated and loaded into the boiling flask. The disaggregation was accomplished by hand with the laboratory operator wearing rubber gloves in order to prevent moisture exchange from the skin onto the soil.

2.) About 100-250 ml of reagent grade toluene was decanted onto the soil. Enough toluene should be used in order to fill the buret stopcock connector to a point above the elbow joint that branches off to the boiling flask. This will allow the toluene to be recycled back into the boiling flask since toluene is less dense than water and

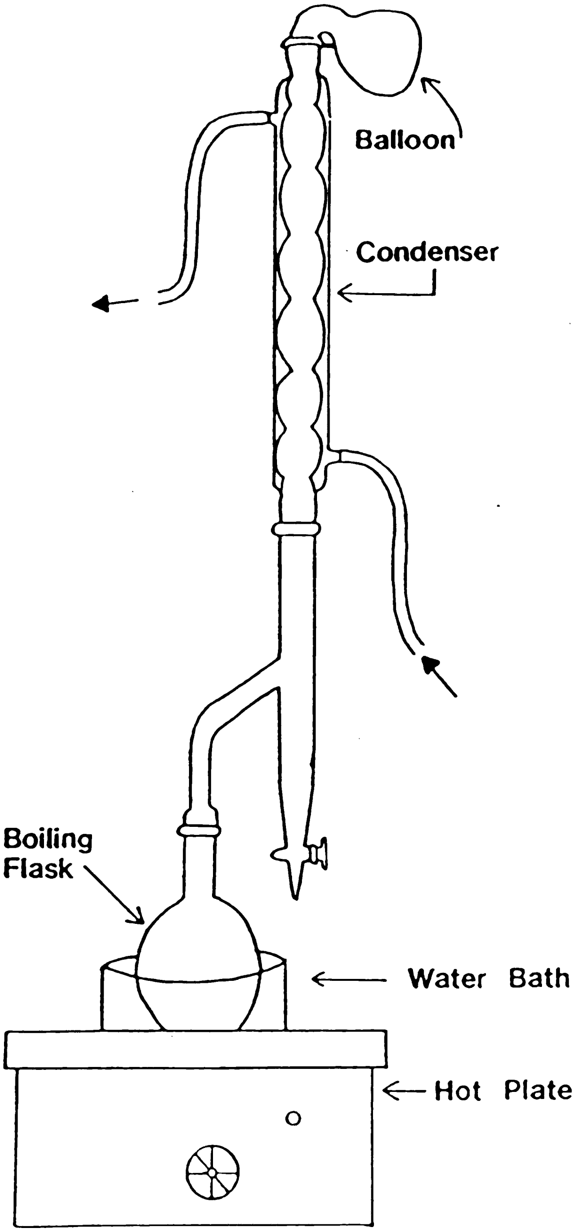


Figure 15. Soil water extraction apparatus (azeotropic distillation).

will therefore float on top of the extracted water. Toluene is used as the azeotrope because it boils at a very low temperature, has a low solubility in water and there is a substantial density difference between toluene and water.

3.) The boiling flask was then connected to the extraction apparatus as shown in figure 15 and the hot plate was turned on and set to an intermediate setting. A water bath was used to help distribute the heat more evenly and to moderate the temperature within the boiling flask. After a few minutes, the toluene began to boil and condensation would begin to form in the bottom of the buret. As the amount of condensation grew, a meniscus would form marking the interface between the water and the toluene.

4.) The initial few drops of extracted water are drained off and discarded in order to flush out the bottom of the buret. The remaining water that condenses at the bottom of the buret is collected by opening the stopcock and allowing the water to drain into a 25 ml glass sample bottle. This may require that the stopcock be opened to drain off the collected water and then closed to allow for more collection several times during the extraction. Care should be taken to keep the meniscus above the stopcock to avoid the transfer of toluene into the sample bottle.

5.) Once the sample bottle is almost full or if there is no more water forming in the bottom of the buret, a few ml of melted paraffin should be poured onto the extracted water. The paraffin will combine and wick off any minor amounts of toluene that may have passed over during the extraction process. It also serves to seal the water sample from the atmosphere. After the paraffin is applied, the sample bottle should be sealed by tightly screwing on its cap. The amount of water extracted varied depending on the soil type and mass. Typically, 1000 gm of moist clay rich soil yielded about 50 ml of extracted water.

APPENDIX C

STABLE ISOTOPE SYSTEMATICS

The stable isotopes ^{18}O and ^{2}H can be employed to distinguish between differing water masses. Due to the different vapor pressures of $H^{18}OH$ and ^{2}HOH, fractionation will occur during evaporation. The amount of fractionation that occurs is dependant on the reaction rate and the temperature (Dansgaard, 1964). Fractionation will increase with increasing reaction rate and decreasing temperature. The affect that temperature has on this relationship allows for the use of ^{18}O and ^{2}H to establish seasonal and large scale climate variations found in the water masses. Therefore, a water mass that formed during the winter or during a time of glaciation would be depleted in heavy isotopes relative to either summer or modern waters.

Deuterium and ^{18}O values are expressed in terms of delta or del units as follows:

Eq.5

$$\text{del sample } (0/00) = (R_{sample} - R_{standard})/R_{standard} * 1000$$

Where R represents the isotopic ratios, $^{18}O/^{16}O$ or $^{2}H/H$, for the sample or standard. The del values are reported in terms of parts per thousand. SMOW (Standard mean ocean water) is the standard which meteoric waters are measured against.

Craig, 1961 found that meteoric waters plotted along a line defined by:

Eq.6 $$\text{del }^{2}H = 8 * \text{del }^{18}O + 10$$

In addition to the Craig meteoric line, researchers in Simco, Ontario have found that precipitation in this area falls along a line defined by (Desaulniers et al, 1981):

Eq.7 $$\text{del }^{2}H = 7.5 * \text{del }^{18}O + 12.6$$

This line can be thought of as a subset of the Craig line that reflects climate conditions that are closer to those found in Michigan. Factors that cause a water mass to plot off the above meteoric lines are exchange with rock minerals, hydration of silicates, H_2S exchange, and evaporation from an open surface (Fritz, 1982; Payne, 1972).

APPENDIX D

TRITIUM SYSTEMATICS

During the mid to late 1950's and early 1960's, a large amount of tritium (^3H) was introduced into the atmosphere via nuclear weapons testing. Peak concentrations were measured in precipitation during this period in excess of 1000 TU's at both the Chicago and Ottawa stations, where 1 TU = 1 tritium atom per 10^{18} hydrogen atoms. Yearly average tritium concentrations were recorded at both stations, which allowed for the generation of a tritium input function (Appendix E). In addition to the tritium produced by nuclear testing, there is constant production of this isotope in the upper atmosphere by the reaction (Vinogradov et al., 1968):

$$\text{Eq.8} \qquad {}^{14}N + \text{neutron} \rightarrow {}^{12}C + {}^{2}H$$

The rate of atmospheric production has been estimated as .25 atoms/cm^2/s (Lal and Peters, 1962). A recent study by Robertson and Cherry (1989, in press), has shown that waters recharged prior to 1953 (i.e. before nuclear testing) would have a tritium concentration of .45 TU or less. Based upon the tritium half life of 12.43 yrs and this value of .45 TU, the natural meteoric level of approximately 3 TU would be expected in rainwater that fell in 1953. A tritium content of less than .45 TU is therefore the cutoff point between "dead" and "bomb" waters.

It is then possible to use tritium to date the rate of water movement through soils and sediments by locating within a soil boring the interface between bomb and pre-bomb waters. Several researchers throughout the United States and Canada have used this relationship to calculate recharge rates (Delcore and Larson, 1987; Delcore, 1985; Dincer et al., 1971, 1974; Larson et al., 1987; Allison and Hughes, 1972, 1975; Andres and Egger, 1985; Atakan and Roether, 1974; Knott and Olimpio, 1986; Rehm et al., 1982; Vogel and Thilo, 1974; and Allison and Holmes, 1973). In addition, tritium has been used by researchers to determine groundwater age (Nir, 1964; von Buttlar, 1958, 1959), storage (Erikson, 1958; Begemann and Libby, 1957), dispersion and advection (Rabinowitz et al., 1977; Egboka et al., 1983; Larson et al., 1987; Green et al., 1972; Robertson and Cherry, in press), and as a groundwater tracer in tills (Hendry et al., 1983, 1986; Hendry, 1988; Grisak and Cherry, 1975; Grisak et al., 1976; Cravens and Ruedisili, 1987; Foster, 1975; Keller et al., 1986; Brown, 1961).

The use of tritium as a groundwater tracer is dependant on it being a conservative tracer. Therefore, it must interact with the clay minerals in the same manner as bulk water and not undergo fractionation as it moves through the sediment. The conservative nature of tritium in clay rich sediments has been demonstrated by Stewart (1972) and Corey and Horton (1968). Both experiments used miscible displacement methods to demonstrate that the breakthrough curves for water tagged with tritium, deuterium and protium were identical and therefore neither isotope was fractionated or retarded relative to the others.

APPENDIX E

TRITIUM INPUT FUNCTIONS

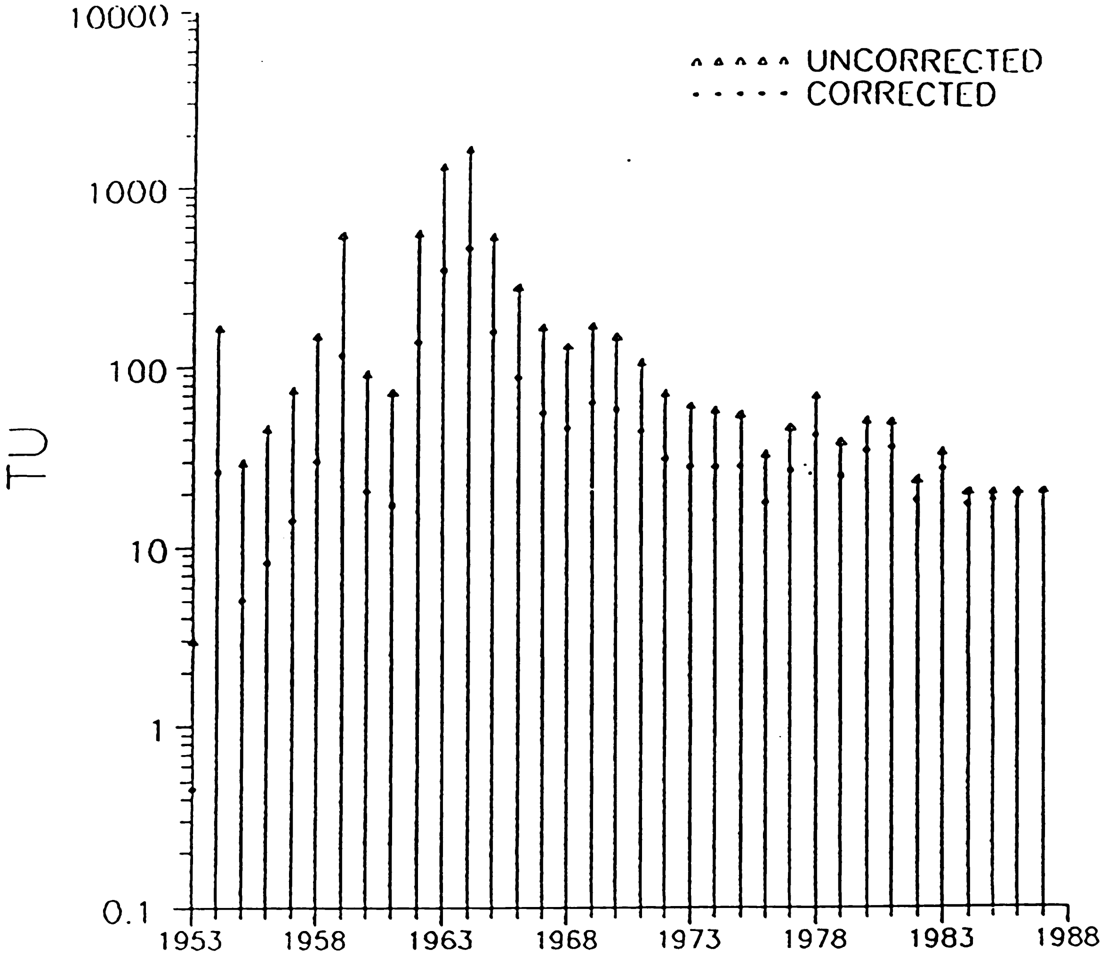


Figure 16. Tritium input function (International Atomic Energy Agency, 1969 - 1986).

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